Development of the Fabrication Technology of Wide Uranium Foils for Mo-99 Irradiation Target by Cooling-roll Casting Method

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Abstract

An alternative fabrication method for polycrystalline uranium foils has been investigated using a cooling-roll casting method at KAERI since 2001, in order to produce a medical isotope ^{99}Mo, the parent nuclide of 99^{m}Tc. The fabrication method of wide uranium foils produced by a cooling-roll casting was recently developed to improve the quality of the uranium foils and the economic efficiency of the foil fabrication with modifications of the casting apparatus and adjustments of the process parameters. A continuous polycrystalline LEU foil with a thickness range of 100 to 150㎛ and a width of about 50 mm, exceeding 5m in length for a batch procedure, could be fabricated with a better quality of the uranium foil and a higher yield of the foil fabrication, through improvements of the casting apparatus and variations of the process parameters.

1. Introduction

Generally, the conventional fabrication method for uranium foils for Mo-99 irradiation target [1-2] has the disadvantages of complicated processes such as the following: casting the uranium; cutting the resulting ingot to a suitable size for a hot rolling; rolling a thick piece of the ingot through many passes to gradually thin it to fabricate a uranium foil with a thickness of about 100 μm; and finally a heat-treatment at ~800 °C and quenching the fabricated uranium foil to produce the required gain size and orientation.

In the conventional method, the uranium must be heated and rolled under a vacuum or in an inert atmosphere because it is a reactive material. A hot rolling is repeated several times to obtain a suitable thickness of the uranium foil. As the hot-rolling process takes a long time, the productivity is relatively low. A washing/drying process must be done to remove the surface impurities after a hot rolling. In order to obtain a fine polycrystalline structure which has a more stable behavior during an irradiation, heat-treatment and quenching must be performed. The high hardness and the low ductility of the uranium make it difficult to roll the foil. The foil is liable to crack due to a residual stress during the process, resulting in a low yield. Hence, it is difficult to fabricate Mo-99 irradiation target in commercial scale by the conventional method.

An alternative fabrication method for polycrystalline uranium foils has been investigated using a cooling-roll casting method at KAERI since 2001 [3], in order to produce a medical isotope ^{99}Mo, the parent nuclide of 99^{m}Tc. Low enrichment uranium foils were fabricated through cooling-roll casting of uranium melt without a hot rolling process and a heat-treatment process. An improvement in productivity and process economic is expected due to process simplification and better quality from the absence of any residual stress on the foil. In the present study, the fabrication method of wide uranium foils produced by a cooling-roll casting has been developed to improve the quality of the uranium foils, and the economic efficiency of the foil fabrication, with modifications of the casting apparatus and adjustments of the process parameters. The wide uranium foils have been obtained through a rapid cooling directly from a melt.

2. Experimental Process

2-1. Modification of the casting apparatus

The casting apparatus of wide uranium foils has been modified to improve the quality of the uranium foils and the economic efficiency of the foil fabrication. A degassing system for the uranium melt was installed in the cooling-roll casting apparatus, in order to reduce the impurities and the numbers of holes in the uranium foils. A discharge control method for the uranium melt was applied to stabilize the fabrication process and to increase the yield of the uranium foils through the prevention of a melt leakage. As uranium has a low thermal conductivity, the collection chamber was modified to soundly fabricate uranium foils without significant defects, which improves the quality and the yield of the uranium foils. In addition, a heating system for the cooling roll was installed to reduce the number of holes in the uranium foils and to improve the surface state of the uranium foils.

2-2. Fabrication of the uranium foils

Uranium lumps (99.9 % pure) were charged and induction-melted in a high-temperature-resistant ceramic nozzle. The dimension and the surface state of the uranium foils were mainly adjusted by the revolution speed of the cooling roll, the ejection pressure of the melt and the superheat of the metal. The superheated molten uranium metal was discharged through a small slot in the nozzle onto a rotating cooling-roll under the condition where the slot was located close to the cooling roll. The uranium foils were rapidly cooled by a contact with the rotating roll driven by an electric motor in an inert atmosphere so that fine crystalline grains of the uranium foil with an irregular orientation are formed. The rapidly solidified foils were collected in a container.

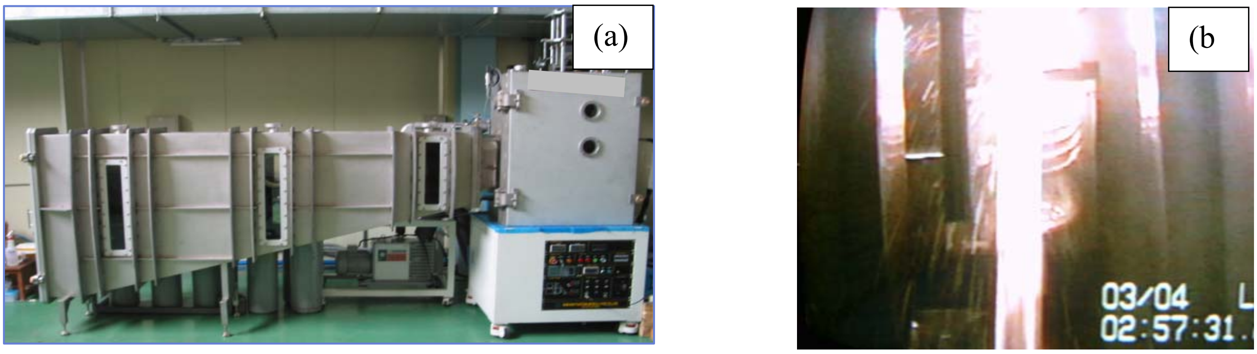


Fig. 1. Experimental apparatus (a) and scene (b) of a cooling-roll casting.

2-3. Analysis of the uranium foils

The dimensions of the uranium foils were measured at several positions along each foil, using a micrometer and vernier-calipers. The surface morphology of the uranium foils was examined with a scanning electron microscope (SEM). The uranium foils were polished to 0.3μm in diamond paste, and a metallographic observation was performed for several sections of the foils, using a scanning electron microscope (SEM). A X-ray diffractometer (XRD) using Cu K_{α} radiation and a Ni filter was used to determine the phase and the preferred orientation for both surfaces of the foils.

3. Results and Discussions

Fig. 1 shows the typical appearance of a continuous foil of 50mm in width with a high flexibility and a good collection state, fabricated by the cooling-roll casting apparatus. The uranium melt was cast to fabricate the uranium foils with a high productivity, in a few seconds by the cooling-roll casting method, which leads to high economical benefits. The uranium foil with a thickness ranging from 100 to 150μm was cast continuously, exceeding 5m in length for one batch. The feeding control method of the uranium melt was applied to stabilize the casting process and to increase the yield of the uranium foils through the prevention of a melt leakage. Wide uranium foils, almost the same width as the nozzle slot, were fabricated by controlling the superheating and the injection pressure of the uranium melt. As uranium has a low thermal conductivity, it is not easy to soundly fabricate uranium foils without wrinkles and indents in the collection process. Hence, the collection chamber was modified to soundly fabricate uranium foils without significant defects, which improves the quality and the yield of the uranium foils.

Fig. 1. Typical appearance of a continuous foil of about 50mm in width with a high flexibility and a good collection state, fabricated by the cooling-roll casting apparatus; (a) DU foil, (b) LEU foil.

It is difficult to fabricate the uranium foils without holes due to pure metal not having coexistent region between soild and liqid. A degassing system for the uranium melt was installed in the cooling-roll casting apparatus, in order to reduce the impurities and the holes of the uranium foils. A heating system for the cooling roll was installed to improve the surface state and to decrease the number of the holes for the uranium foils. Then, uranium foils were soundly fabricated, through the installation of the degassing system and the heating system. The uranium foils showed a good surface state and very small holes, as shown in Fig. 2.

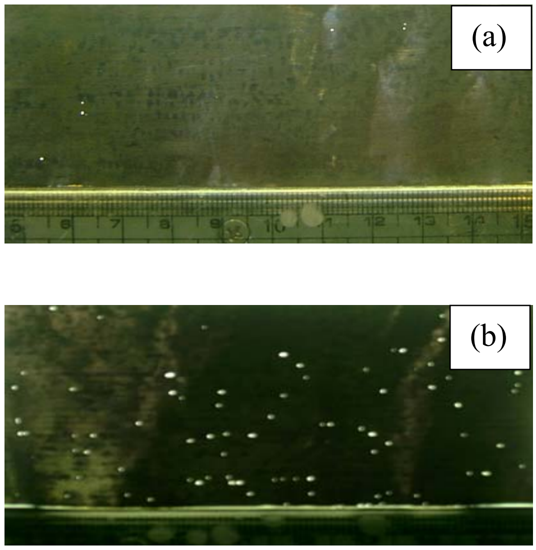


Fig. 2. Uranium foil fabricated by the degassing of the melt and the heating of the cooling-roll; (a) DU foil, (b) LEU foil.

The thickness of the uranium foils was mainly adjusted by the revolution speed of the cooling roll and the ejection pressure of the melt. As the revolution speed increased, the thickness of the uranium foils decreased; however, as the injection pressure increased, the thickness of the uranium foils increased, as shown in Fig. 3. The thickness of the uranium foils was mainly influenced by the revolution speed of the cooling roll and the ejection pressure of the melt.

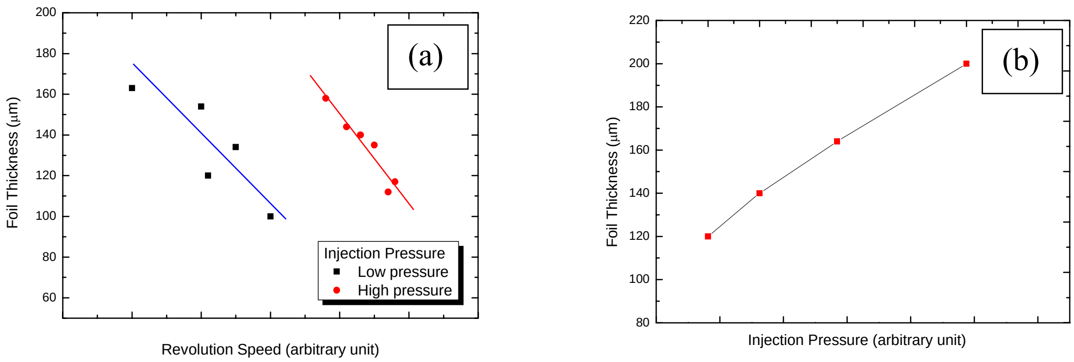


Fig. 3. The effect of the revolution speed (a) and the injection speed (b) on the thickness of the uranium foils.

Figs. 4 ~ 5 show the scanning electron micrographs for the free surface and the wheel-side surface the obtained LEU foil, respectively. Both surfaces of the LEU foil have a relatively good surface state, not having cracks and impurities. The wheel-side surface of the LEU foil like the roll surface was much smoother than the free surface of the LEU foil.

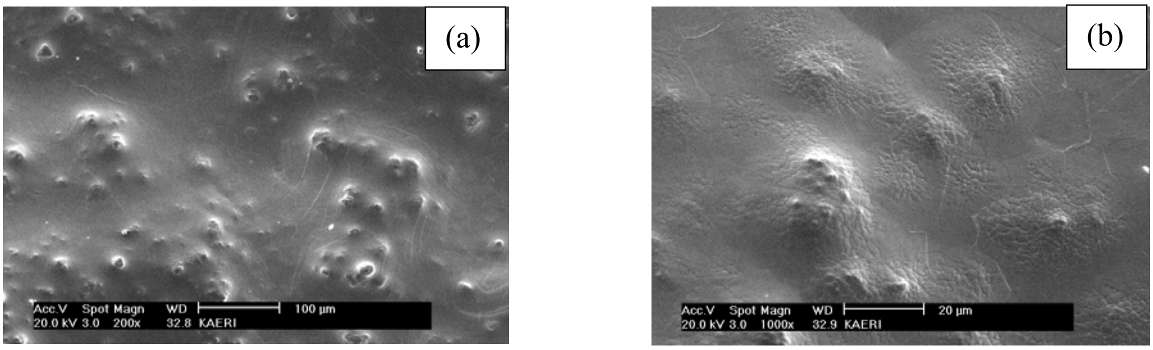


Fig. 4. Scanning electron micrographs of the free surface of the obtained LEU foil; (a) x200, (b) x1000.

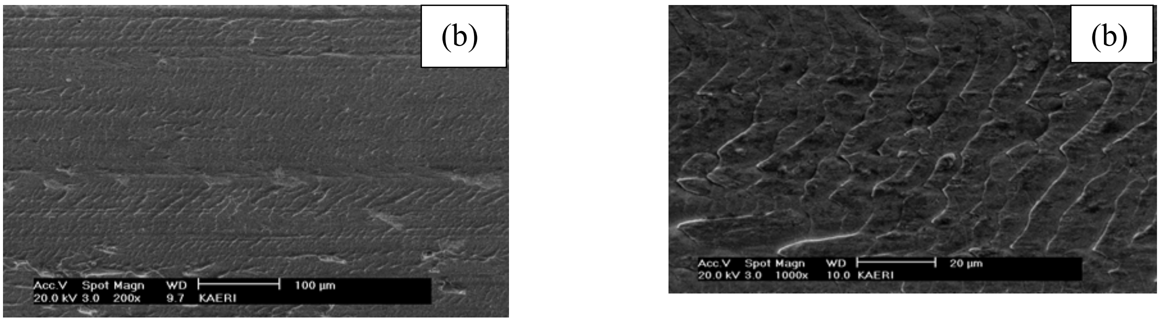
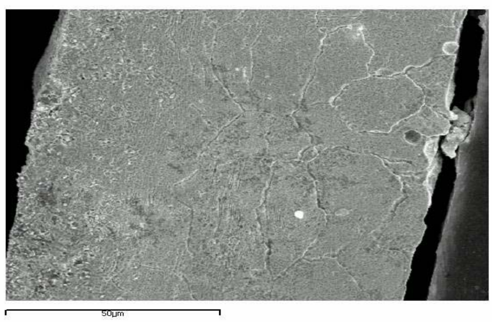


Fig. 5. Scanning electron micrographs of the wheel-side surface of the obtained LEU foil; (a) x200, (b) x1000.

Fig. 6 shows the scanning electron micrograph for cross-sections of the obtained LEU foil. The LEU foil showed a difference in grain size according the location of the foil with fine grains below about 30 microns in size. The grain size of the LEU foil increased, as the distance from wheel-side surface increased. Fig. 7 shows the X-ray diffraction patterns of the obtained foils. The phases of the rapidly solidified foil are found to be the α-U (orthorhombic) phase. Hence, it is not necessary to heat-treat the hot-rolled foil and quench it from about 800 °C to form fine grains, as the uranium foil having fine grains is directly obtained by the rapid solidification effect. It is expected to be able to prevent the uranium foils from an excessive swelling by the an-isotropic growth behavior during an irradiation.

Wheel-side surface



Free surface

Fig. 6. Scanning electron micrographs for cross-sections of the obtained LEU foils.

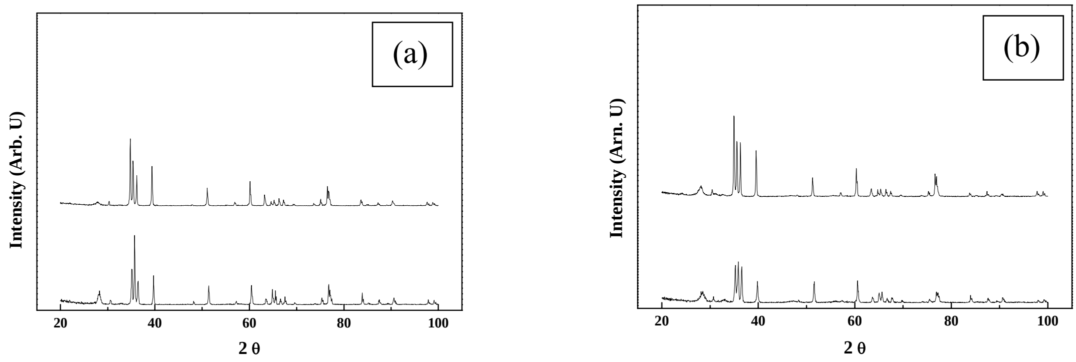


Fig. 7. X-ray diffraction patterns of uranium foils; (a) free surface, (b) wheel-contact side surface.

4. Conclusion

1) The fabrication method of wide uranium foils produced by a cooling-roll casting was developed to improve the quality of the uranium foils and the economic efficiency of the foil fabrication with modifications of the casting apparatus and adjustments of the process parameters.
2) A continuous polycrystalline LEU foil with a thickness range of 100 to 150㎛ and a width of about 50 mm was fabricated, exceeding 5m in length for a batch procedure, with a better quality of uranium foil and a higher yield.
3) The thickness of the LEU foil was mainly controlled by the revolution speed of the cooling roll and the ejection pressure of the melt.
4) The LEU foil had a relatively good roughness on the surface, with few impurities. The wheel-side surface of the LEU foil was rather smoother than the free surface of the LEU foil.
5) The LEU foil showed a difference in grain size according to the location of the foil, but fine grains below about 30 microns in size.

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Reference

[1] J. L. Snelgorve, et al., Proc. 22 nd International Meeting on Reduced Enrichment for Research and Test Reactors, Budapest, Tennessee, Hungary, Oct. 3-8, 1999.
[2] G. F. Vandegraft, et al., Proc. 22 nd International Meeting on Reduced Enrichment for Research and Test Reactors, Budapest, Tennessee, Hungary, Oct. 3-8, 1999.
[3] K. H. Kim, Hee-Jun Kwon, Jong-Man Park, Yoon-Sang Lee, and Chang-Kyu Kim, Journal of Korean Nuclear Society, Vol. 33, No. 4, pp. 365-374, Aug. 2001.